

Mobilization and Transport of Soil Particles during Infiltration Experiments in an Agricultural Field, Shenandoah Valley, Virginia

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Evidence that fine particles mobilized and transported in soils and aquifers can have a profound influence on contaminant migration has spawned much interest recently in understanding colloid transport in natural materials. Repeated infiltration experiments on an initially dry field soil were conducted to evaluate rates of mobilization of fine particles over time and to investigate the importance of transient-flow events on particle transport. Water flow was measured in zero-tension lysimeters at 25 cm depth. For repeated infiltration events and for all plots, water flow sharply increased shortly after initial ponding of water at the soil surface, maintained a relatively steady level during the period of ponding, and decreased gradually thereafter. Particle concentrations measured in the pan lysimeters ranged from 7 mg L⁻¹ to 265 mg L⁻¹ and were typically on the order of 10 to 100 mg L⁻¹. Greatest particle mass flux was observed during the initial infiltration experiment on each plot. During four subsequent infiltration experiments, all conducted within 250 min of the first event, steady mass fluxes were observed that were approximately 70% of the average value seen in the first flush of water through a dry soil, indicating that the supply of mobile soil particles is only sparingly reduced over closely spaced infiltration events. All peak particle concentrations and mass fluxes occurred near either the rising limb or the falling limb of the water flux hydrograph, presumably reflecting the movement of air–water interfaces during imbibition and drainage.

Introduction

Evidence that fine particles mobilized and transported in soils and aquifers can have a profound influence on contaminant migration has spawned much interest recently in understanding colloid transport in natural materials. Particle mobilization and transport in soils is directly

controlled by solution chemical composition (1–5) as well as rates of water movement through soil (6–8). Despite considerable previous work using columns of soil in the laboratory and limited field experimentation, questions remain about the importance of rapid temporal variations in both flow rates and chemical composition of infiltrating water on the mobilization and transport of soil fines in the vadose zone.

Kaplan et al. (3) observed that particle concentrations eluted from repacked lysimeters were directly related to the kinetic energy of water movement. Conversely, Ryan et al. (8) did not observe any correlation between water application rates and mobilized particle concentrations measured in field-installed lysimeters. In the same study, Ryan et al. (8) observed diminished particle concentrations after a series of simulated rainfall events. They suggested that particle regeneration rates are dependent on wetting–drying cycles and biological activity in soils. Similarly, Jacobsen et al. (7) observed no correlation between particle concentrations collected and water application rates in intact soil cores.

Changes in chemical composition of interstitial waters caused by wetting and drying of a soil can exert a very strong influence on particle mobilization (e.g., ref 6). Furthermore, drying of soil leads to creation of air–water interfaces to which particles adhere strongly (e.g., refs 9–12) and can lead to clay structures from which the water eventually withdraws (13). Upon rehydration by chemically dilute infiltrating water, the particle aggregates can be destabilized and the resulting fine particles mobilized. If the air phase is static, the particles will be held in the soil. Wan and Wilson (10), using flow visualization in a micromodel, observed strong sorption of hydrophobic and hydrophilic particles to the air–water interface. This sorption process is fast and irreversible, suggesting that significant immobilization of particles can occur in soils with a stationary air phase. Even under steady-flow conditions, however, air bubbles may move and carry along the associated fine particles. This phenomenon is the basis for the flotation process used in the minerals industry, for example (14). Under the transient-flow conditions caused by infiltration of water into a soil, a reasonable expectation is for enhanced particle transport along with the wetting and drying fronts. Goldenberg et al. (9) measured particle concentrations from 20 to 150 times larger at bubble interfaces relative to bulk water in a micromodel, implying that the movement of bubbles would enhance particle transport rates. Results from a mathematical model that mimics strong adsorption of particles to air–water interfaces (by making adsorption a function of moisture content) confirms that relatively high particle concentrations may be associated with a wetting front (15).

The available evidence motivates two questions. First, is the source of soil colloids depleted after an initial “flush” of infiltrating water, presumably due to either increased flow or a decrease in ionic strength of the soil water or both? If soil particles are rapidly depleted, the effect of colloid-facilitated transport in soils may be minor because of a limited supply of soil fines, or the effect may be important only after prolonged periods of dry weather during which biological and physical processes in the soil have enough time to generate a supply of fine particles. Second, is the transport of fine particles during flow transients (presumably caused by movement of air–water interfaces) a process of practical consequence? Although Goldenberg et al. (9) showed a one- to two-order-of-magnitude increase in concentration of colloids near bubbles, no results are available to gauge the importance of this observation in the field. We conducted

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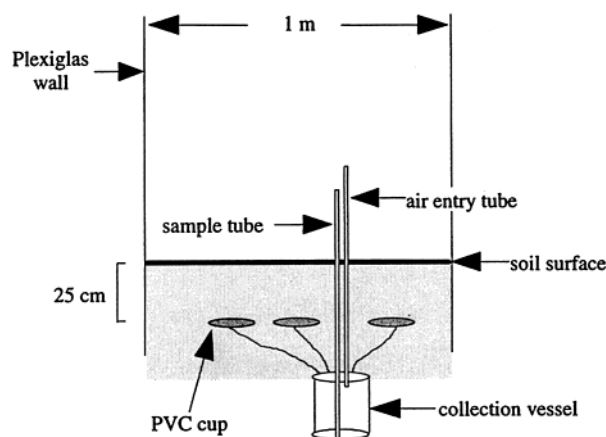


FIGURE 1. A schematic of the installation for each plot. The Plexiglas box is 60 cm in the third dimension.

a set of infiltration experiments in an agricultural field to address these two questions.

Methods

Site Description and Field Methods. Our field site is located in the Shenandoah Valley of Virginia. Soils are classified as Frederick and Lodi silt loam (Typic Hapludult) which are characteristically highly structured, deep and well drained having formed from the residuum of extensively weathered limestone (16). The soil is 24% clay, 34% silt and 40% sand with organic carbon concentrations ranging from 1 to 2% in the upper 10 cm of soil to less than 1% from 10 to 30 cm (17).

A transect of twelve zero-tension lysimeters was installed at 25 cm depth during the summer of 1997 (18) along the edge of a field used primarily for no-till corn production. Spaced approximately 5 m apart on a relatively flat slope, each lysimeter was constructed of three cylindrical, 15.2-cm-diameter PVC cups connected by reinforced Tygon tubing to a collection vessel buried in a soil trench next to and lower than the collection area. The cups were filled with 0.75-mm-diameter clean quartz sand and were placed into hand-excavated cavities with the tops pressed tightly against the undisturbed overlying soil. The total collection area for each lysimeter was 547 cm². Air entry/exit and sample tubes connected the collection vessel to the surface of the soil. For this study, four lysimeter plots were selected from the earlier installation: Plots 1, 4, 6, and 11 (numbered by Sprague (18)). A 30-cm-deep trench was dug along each side of the rectangular (1 m × 0.6 m) plots. A preconstructed Plexiglas sleeve was placed over the soil block leaving 76-cm-high walls extended above the soil surface (Figure 1). The walls were sealed to the soil with a concrete-bentonite mix. In late June of 1998, one month prior to experimentation, the vegetation was cut to a uniform height of 2 cm, the trimmings were removed, and the soil surface was covered evenly with 2 cm of cow manure mixed with wood chips, simulating what is applied to the field by the farmer.

Repeated infiltration experiments were conducted on each plot. Immediately before the first application of water, 1.7 cm of water was applied as a broadcast spray. Using the sample tubes, the collection vessels for the lysimeters were pumped out and rinsed with added deionized water until no turbidity was observed, thereby cleaning out any accumulated soil water and particles from previous rain events on the exposed plots. The soil surface within the Plexiglas enclosure was then covered with a fine mesh screening to minimize disturbance of the soil surface during the experiments. A large plastic tarp was then placed on the plot and draped up over the walls of the Plexiglas reservoir. A 0.1 mM NaCl solution was poured into the enclosure on top of the plastic

tarp to a depth of 10 cm. The tarp was removed rapidly to allow water to contact the soil surface and thus to begin the experiment. A peristaltic pump attached to the sample tube from the collection vessel was turned on, collecting water that drained by gravity into the collection vessel. The end of the tube was placed in a graduated cylinder, and water volume was monitored continuously. Water samples were collected at intervals ranging from 30 s to greater than 10 min, depending on flow rate, with longer-interval sampling occurring as flow rate decreased toward the end of an experiment. The infiltration experiment was considered complete when no water was collected in the vessel for approximately 15 min. Subsequent experiments were performed following the same protocol. The repeated infiltration experiments were conducted by sequentially ponding water at initial depths of 10, 5, 10, 20, and 5 cm in the Plexiglas enclosure. The duration of the entire sequence of infiltration experiments ranged from 174 to 252 min among the four plots.

Particle Concentration. Particle concentration was determined spectrophotometrically at 400 nm. A standard curve was developed from successive 50% dilutions of a stock soil suspension. Approximately 40 g soil from the site was stirred in a 2-L flask containing 5 mM NaCl solution on a magnetic plate for 1 h. The soil suspension was allowed to settle for 2 h at which time the particles remaining in suspension are less than 2.0 μm in diameter (by Stokes Law). A known volume of suspension was passed through a preweighed 0.2 μm filter, and the mass of particles retained on the filter was measured. A previous analysis of particles eluted from intact columns of the same soil subjected to infiltration experiments in the laboratory by Kauffman (17) demonstrated that the average size of soil particles was 253–270 nm with a low average polydispersity (0.34), indicating that the majority of particles mobilized from these soils can be considered colloidal.

Particle mass flux rates were obtained by multiplying measured flow rates by measured particle concentrations. The volume-weighted average particle concentration for each infiltration experiment was calculated by dividing the total mass flux of particles (discharge multiplied by concentration) by the total discharge.

Results

Water flow rates increased rapidly after initiation of ponding on each plot, reaching peak flow within 30 to 60 s (Figure 2). Flow continued at a relatively high rate over the duration of ponding and then decreased gradually. Flow rates throughout the duration of ponding on the four plots ranged from about 500 to 1200 mL min⁻¹ over the sequence of water-addition experiments. The peak flow rates on individual plots were reasonably close across the sequence of infiltration experiments suggesting that flow rates were not affected strongly by the depth of ponded water and that the soil conductivity did not change appreciably.

The particle concentrations during the infiltration experiments ranged from 17 to 147 mg L⁻¹ for Plot 1, 7 to 119 mg L⁻¹ for Plot 4, 30 to 265 mg L⁻¹ for Plot 6, and 11 to 62 mg L⁻¹ for Plot 11 (Figure 2). Volume-weighted average concentrations also varied across water-addition events for individual plots, varying in Plot 4, for example, from 44.1 mg L⁻¹ during the first 5-cm event to 16.7 mg L⁻¹ during the 20-cm event, then increasing to 30.2 mg L⁻¹ during the second 5-cm event (Table 1). Considering all four plots, the volume-weighted average particle concentrations decreased from the initial 10-cm infiltration event to the 20-cm event. For Plots 4 and 6, the volume-weighted average concentrations increased during the last 5-cm event. The decrease in the average concentration between the initial 10-cm event and the subsequent 10-cm event was approximately 25% for all the plots.

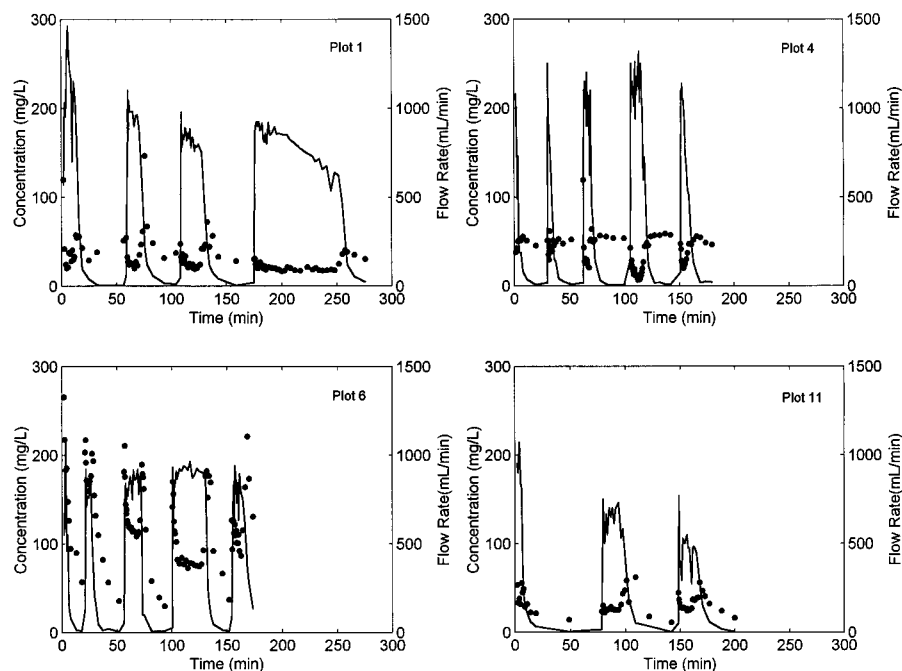


FIGURE 2. Particle concentrations and water flow rates in each plot. The solid circles (••) represent particle concentrations, and the lines illustrate flow rates which were sampled at the same times as the particle concentrations. In each panel, the sequential experiments for each plot are shown in the order of 10-cm, 5-cm, 10-cm, 20-cm, and 5-cm pond heights. For Plot 1, the final 5-cm experiment was not conducted, and, for Plot 11, neither the 20-cm nor the final 5-cm experiment was conducted. Note that axes are identical for all four panels.

TABLE 1. Volume-Weighted Average Particle Concentration (mg L^{-1}) for Each Infiltration Experiment

pond height (cm)	plot 1	plot 4	plot 6	plot 11
10	38.2	43.8	192.4	41.9
5	31.8	44.1	178.5	30.3
10	28.2	34.2	133.6	32.5
20	21.5	16.7	101.9	
5		30.2	117.3	

The slope of the curve relating cumulative particle mass collected to cumulative water volume collected is indicative of particle production (Figure 3). The curves slope relatively steeply at first, but after the presaturation ponding, the slope of these curves remains relatively constant for individual plots indicating that particle generation rates were relatively constant with time.

Particle concentrations for Plots 1 and 6, and less clearly for Plot 11, were highest on the ascending and descending limbs of the hydrograph (Figure 2). The particle mass flux, calculated as the product of measured concentration and measured flow rate, peaked during the rising and falling limbs of the hydrograph for all plots (Figure 4). The peaks can be seen more easily on plots for individual infiltration events (Figure 5), and these peaks were evident for all of the experiments with the exception of the initial 5-cm event.

Discussion

Volume-weighted average particle concentrations were highest during the first 10-cm infiltration experiment, decreased for subsequent infiltration events in general, and increased during the final 5-cm infiltration event on Plots 4 and 6 (Table 1). The slopes of the straight-line segments relating cumulative particle flux to cumulative water flux after the initial ponding (Figure 4) indicate production rates of from 23 mg to 104 mg of particles per L of water throughflow. These results show that, after an initial decrease in the rate of particle production, a relatively steady generation rate of soil particles is achieved, even for quantities of

infiltrated water equivalent to the expected amount of precipitation for six months at our site. These field results are consistent with those from experiments on intact soil cores from this same field, which showed nearly constant rates of production of fine particles over extended periods of flow through the cores (17). A fairly continuous rate of particle supply also is indicated from other work. Ryan et al. (8) observed an initial decrease in the mean particle concentrations collected in zero-tension lysimeters during simulated rainfall applications to a field plot following an initial simulated rainfall event but continued to collect particles at an average concentration of several hundred mg L^{-1} during subsequent sprinkling experiments. Thus, although Ryan et al. (1998) concluded that the time interval between their simulations was shorter than the time periods needed for particle regeneration (e.g., soil drying, earthworm activity), their results suggest that particles were generated at a fairly high rate even after a total infiltration of 25 cm of water.

Concentrations of fine particles in the range of 10's to 100's of mg L^{-1} , such as we observe (Figure 2), appear to be typical. As mentioned above, Ryan et al. (8) report particle concentrations of hundreds of mg L^{-1} . Nightengale and Bianchi (19) report mean concentrations of $\sim 10 \text{ mg L}^{-1}$ in recharge experiments at rates of 16 to 23 cm day^{-1} . The particles, generated in the recharge of water of lower ionic strength than the groundwater at the site, were transported to the water table and thence to producing wells. Concentrations of colloidal-sized particles in the 10's to 100's of mg L^{-1} range not only result in noticeable turbidity of groundwater but also can contribute to facilitation of transport of strongly sorbing materials (e.g., refs 20 and 21).

The calculated particle generation rates may be used to determine the rates of eluviation (translocation of soil particles) through the near-surface soil profile. At the relatively high flow rates induced by ponding water at the surface in our experiments, particle concentrations of about 25 to 100 mg L^{-1} are realistic (Figure 2). For experiments on an intact core from our field site run at soil-water tensions

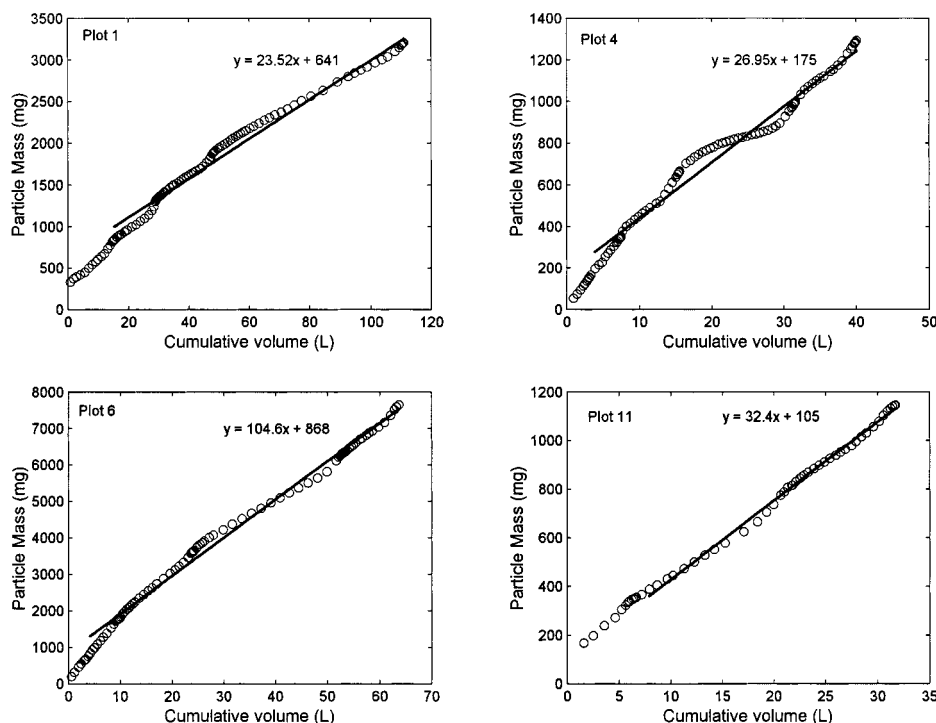


FIGURE 3. Cumulative particle mass collected versus cumulative water volume collected in each plot. The open circles (o) represent the cumulative mass, and the line shows a least-squares fit to the data collected after the initial 10-cm infiltration. The slope of the least-squares line gives particle generation rates, in mg particles per L of water.

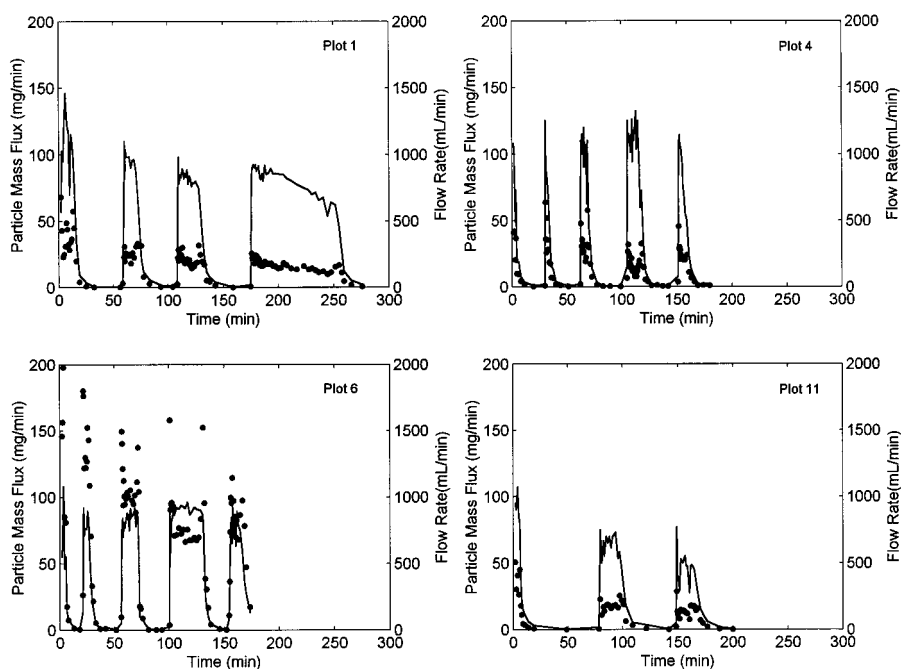


FIGURE 4. Particle mass fluxes and measured water flow rates in each plot. The solid circles (•) represent particle mass fluxes, and the lines illustrate the flow rates which were sampled at the same times as the particle concentrations. In each panel, the sequential experiments for each plot are shown in the order of 10-cm, 5-cm, 10-cm, 20-cm, and 5-cm pond heights. For Plot 1, the final 5-cm experiment was not conducted, and, for Plot 11, neither the 20-cm nor the final 5-cm experiment was conducted. Note that axes are identical for all four panels.

of -8 and -19 cm, effluent colloid concentrations were 110 mg L^{-1} and 10 mg L^{-1} , respectively (22). Thus, a range of 10 mg L^{-1} to 100 mg L^{-1} for concentrations provides sensible estimates under a variety of conditions. For each meter of water that infiltrates to the 25-cm depth (the annual precipitation for this site is about 90 cm), we can infer that from 10 to 100 g of particles are transported per m^2 of soil surface depending on whether the average particle concen-

tration is taken as 10 mg L^{-1} or 100 mg L^{-1} . If the bulk density of the soil is taken to be 1400 kg m^{-3} , the total mass of soil above the 25-cm depth (per m^2 of soil surface) is 350 kg. Thus, the near-surface rates of particle translocation at our site indicate time scales for eluviation on the order of 10^3 years. These rates are unlikely to be maintained at greater depths, however. For example, Kretzschmar et al. (23) report high filter coefficients in saprolite cores and conclude that

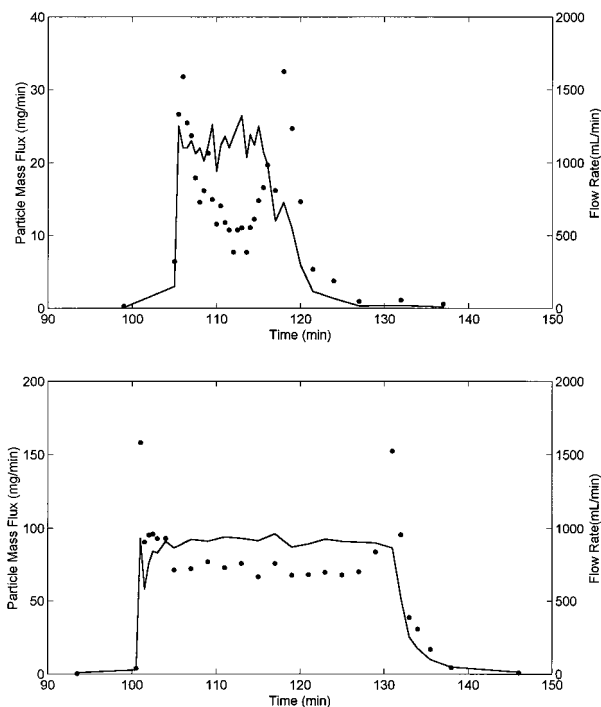


FIGURE 5. Particle mass fluxes and measured water flow rates for the 20-cm infiltration event in Plots 4 and 6. The solid circles (—•) represent particle mass fluxes, and the lines represent the flow rates. These two individual experiments are illustrative examples selected from the entire set of results shown in Figure 4.

transport of colloidal particles is unlikely through the CB horizon except in cracks or other large openings.

Peaks in particle concentration and in particle mass flux were observed near the rising and falling limbs of the water flux hydrographs in all the plots (Figures 2 and 4). Peak particle concentrations were from 2 to 6 times higher than plateau concentrations (Figure 2), and peak particle fluxes were about a factor of 2 higher (Figures 4 and 5). Data from lysimeter 1 of Kaplan et al. (6) also show higher particle concentrations on the limbs of the water flux hydrograph. During these periods of large changes in flow rates in particular, considerable movement of air–water interfaces is likely to occur due to changes in water saturation. Air–water interfaces have been demonstrated to sorb irreversibly hydrophilic and hydrophobic particles in porous media (9–11). In model porous media, Goldenberg et al. (9) measured clay–colloid concentrations on air bubbles one to 2 orders of magnitude larger than colloid concentrations in the bulk water phase. Our data are consistent with the hypothesis that during imbibition and drainage during and following infiltration events in the field, i.e. the transient portions of the flow, a significant fraction of particles is transported on mobile air–water interfaces.

Data for colloid transport through the vadose zone, particularly under conditions of unsteady flow, are scarce. Bennett et al. (24) state the following: “although some capability to simulate transient flow events exists, there are no experimental studies of transient flow in partially saturated

porous media against which the models of colloid transport can be compared”. The data presented here suggest that processes considered in filtration models such as that of Corapcioglu and Choi (15) may be appropriate, but more data from both laboratory and field systems will be needed to evaluate models and to determine their limitations.

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Literature Cited

- (1) Kretzschmar, R.; Robarge, W. P.; Weed, S. B. *Soil Sci. Soc. Am. J.* **1993**, *57*, 1277–1283.
- (2) Ryan, J. N.; Gschwend, P. M. *Environ. Sci. Technol.* **1994**, *28*, 1717–1726.
- (3) Kaplan, D. I.; Sumner, M. E.; Bertsch, P. M.; Adriano, D. C. *Soil Sci. Soc. Am. J.* **1996**, *60*, 269–274.
- (4) Roy, S. B.; Dzombak, D. A. *Environ. Sci. Technol.* **1997**, *31*, 656–664.
- (5) Kretzschmar, R.; Borkovec, M.; Grolimund, D.; Elimelech, M. *Adv. Agronomy* **1999**, *66*, 121–193.
- (6) Kaplan, D. I.; Bertsch, P. M.; Adriano, D. C.; Miller, W. P. *Environ. Sci. Technol.* **1993**, *27*, 1193–1200.
- (7) Jacobsen, O. H.; Moldrup, P.; Larsen, C.; Konnerup, L.; Petersen, L. W. *J. Hydrol.* **1997**, *196*, 185–203.
- (8) Ryan, J. N.; Illangasekare, T. H.; Litaor, M. I.; Shannon, R. *Environ. Sci. Technol.* **1998**, *32*, 476–482.
- (9) Goldenberg, L. C.; Hutcheon, I.; Wardlow, N. *Transport in Porous Media* **1989**, *4*, 129–145.
- (10) Wan, J.; Wilson, J. L. *Water Resour. Res.* **1994a**, *30*, 857–864.
- (11) Wan, J.; Wilson, J. L. *Water Resour. Res.* **1994b**, *30*, 11–23.
- (12) Powelson, D. K.; Mills, A. L. *Appl. Environ. Microbiol.* **1996**, *62*, 2593–2597.
- (13) Goldenberg, L. C.; Hutcheon, I.; Melloul, A. *J. Hydrol.* **1992**, *138*, 53–75.
- (14) Schulze, H. J. *Physicochemical elementary processes in flotation*; Elsevier: New York, 1983.
- (15) Corapcioglu, M. Y.; Choi, H. *Water Resour. Res.* **1996**, *32*, 3437–3449.
- (16) Hockman, J. R.; Neal, C. F.; Racey, D. L.; Wagner, D. F. *Soil Survey of Rockingham County*; U.S. Department of Agriculture, 1982.
- (17) Kauffman, S. J. Ph.D. Dissertation, University of Virginia, 1998.
- (18) Sprague, L. A. M.S. Thesis, University of Virginia, 1998.
- (19) Nightengale, H. I.; Bianchi, W. C. *Ground Water* **1977**, *15*, 146–152.
- (20) Vinten, A. J. A.; Yaron, B.; Nye, P. H. *J. Agric. Food Chem.* **1983**, *31*, 662–664.
- (21) Saiters, J. E.; Hornberger, G. M. *Water Resour. Res.* **1996**, *32*, 33–41.
- (22) Kauffman, S. J.; Bolster, C. H.; Hornberger, G. M.; Herman, J. S.; Mills, A. L. *Environ. Sci. Technol.* **1998**, *32*, 3137–3141.
- (23) Kretzschmar, R.; Robarge, W. P.; Amoozegar, A. *Water Resour. Res.* **1995**, *31*, 435–445.
- (24) Bennett, D. G.; Crawford, M. B.; Wickham, S. M. *Colloids in saturated and partially saturated porous media: Approaches to the treatment of colloids in Yucca Mountain total system performance assessment*; Final Report, TR-112135, April 1999; Electric Power Research Institute: Palo Alto, CA, 1999.

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